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ENTROPY AND THE HOMOGENEITY
OF SOLID MIXTURES

BY A. A. CAMPOLATTARO, O. H. DENGEL, AND P. A. WALTER

RESEARCH AND TECHNOLOGY DEPARTMENT

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There is a surge of efforts to develop continuous processing techniques for the manufacture of explosives and propellants. A standardized procedure to measure the homogeneity of mixtures produced is required. This paper proposes a new algorithm, based on the concept of entropy, for the calculation of homogeneity of solid mixtures which could be adopted as an industry standard by the scientific community.

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FOREWORD

This work was performed in support of Task No. 2003 - Pilot Plant Scale-Up and Processing Techniques of the IMAD High-Explosives Project.

The algorithm, presented in this report, is ideally suited for the evaluation of the homogeneity of solid mixtures of explosives and propellants produced in batch or twin-screw mixers.

Approved by:

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KURT F. MUELLER, Head
Energetic Materials Division

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INTRODUCTION

When we recently became involved in the study of solid mixing, we knew we were invading an ancient field. This field may reach back as far as the mixing of pigments for the frescoes of Laseaux in Paleolithic times.

The article by S. S. Weidenbaum,[1] published in 1958, with 73 outstanding references provides an excellent review of the subject matter. However, during the subsequent three decades the number of papers, books, and transactions at national and international conferences has reached such a volume that a comprehensive technology review must remain the subject of a separate article. The reader is advised to consult the list of references as a source for detailed information.[2-19]

In the mixing process, two aspects are predominantly important: (1) the actual mechanics of mixing with its machinery and underlying complex theory and (2) the quality control of the homogeneity or goodness of mixtures. In line of principle, a comprehensive theory of mixing would provide information about the goodness of mixtures. However, at present, such a theory does not exist for mixing of solids, at least not at the level of sophistication which is required to predict homogeneity. Regarding characterization techniques and experimental measurements of the goodness of mixtures of solid particles imbedded in polymer matrices, we would like to quote Rauwendaal[2] (p. 323): "Some of the characterization techniques are quite sophisticated and most are very time consuming. Quantitative characterization is very important to workers doing research on mixing. However, such techniques are often impractical in actual polymer processing operations. Visual observation, although qualitative, is often sufficiently accurate to determine whether a product is acceptable or not."

Historically, the determination of the homogeneity or the goodness of mixtures is based on a statistical approach whereby samples are taken at random from a batch of material. The papers of P. V. Danckwerts[17] and A. M. Scott and T. Bridgewater[18] describe the statistical procedures comprehensively. The determination of the homogeneity of, e.g., energetic materials using conventional statistical methods, requires expensive chemical analysis of a large number of samples taken at random from a single batch in order to achieve acceptable error limits. The advent of computer based image analysis systems offers the possibility of automated determination of the distribution of the constituents of a thin sample layer drawn from a batch.

This paper introduces the "anthropomorphic" concept of entropy for the determination of the goodness of mixtures.[20] As will be shown, the entropy concept is ideally suited for integration with computerized image analysis systems. The concept of entropy has found, outside the realm of thermo-dynamics, useful applications in quantum mechanics by J. von Neumann.[21] in information theory by C. E. Shannon and W. Weaver.[22] and in several other fields.[23-25] We hope that the ideas developed in this paper will contribute to a scientific and quantitative approach to the problem of specification of the goodness of solid mixtures.

PARTICLE DISTRIBUTIONS OF SAMPLE LAYERS

A sample is drawn at a random position from the batch of a solid-filled polymeric mixture. The sample size should be large in comparison with the dimensions of the solid particles of the mixture. A rectangular layer is cut out of the sample with the help of a microtome or some other means. The layer should be at least twice as thick as the largest particle class contained in the mixture. The layer is inspected by eye or preferably by an image analyzer and the location of all solid particles of a specified class is determined. Figure 1 depicts a computer generated representation of a hypothetical particle distribution. It has been prepared by graphically assigning 200 points to pairs of random numbers.

ENTROPY OF A POINT DISTRIBUTION

Following the procedure described above, a sample layer is reduced to a set of N points located in a rectangle with sides a and b. Then, a set of coordinates is selected in such a way that the vertices A, B, C, and D of the rectangle are given by A(0,0). B(a,0), C(a,b) and D(0,b).

To specify the degree of order of this set of N points, we apply the concept of entropy. We cover the rectangular set of N points, representative of the sample, with a lattice comprised of n x m cells, where n and m are two positive integers, by dividing the side a into n parts and the side b into m parts. Each cell is identified by two indices (i,j) with i = 1....n, and j = 1....m. The point P(x,y) is considered to belong to the cell (i,j) if its coordinates satisfy the inequalities:

$$(i-1) \frac{a}{n} \leq x < i \frac{a}{n} \qquad i = 1....n-1 \qquad (1)$$

$$(j-1) \frac{b}{m} \leq y < j \frac{b}{m} \qquad j = 1....m-1 \qquad (2)$$

For i = n and/or j = m, the open inequalities become closed. If we define n_{ij} as the number of points enclosed in the cell (i,j), we have
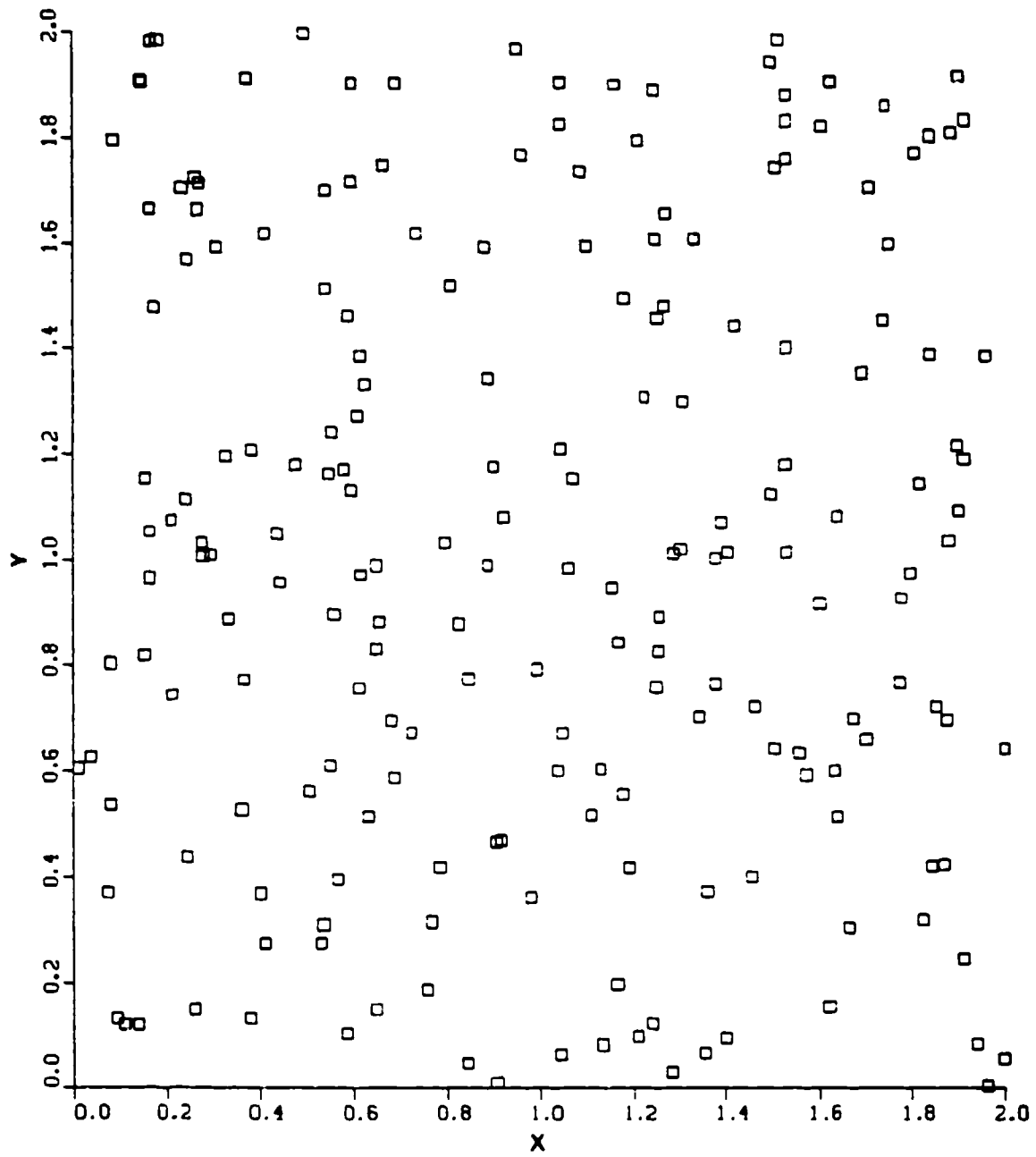
$$\sum_{i,j} n_{ij} = N \qquad (3)$$

FIGURE 1. RANDOM DISTRIBUTION OF 200 POINTS

where the sum is understood to comprise all values of $i = 1....n$ and $j = 1....m$. The frequencies P_{ij} will be defined by

$$P_{ij} = \frac{n_{ij}}{N} \qquad (4)$$

The entropy $S_{n,m}$ associated with the covering lattice of $n \times m$ cells is, thus, defined by us for two dimensional applications:

$$S_{n,m} = -\sum_{i,j} P_{ij} \ln P_{ij}. \qquad (5)$$

Since

$$\lim_{x \to 0} x \ln x = 0, \qquad (6)$$

for a cell (k,h) which contains no points, one has

$$P_{k,h} \ln P_{k,h} = 0. \qquad (7)$$

Because $\ln 1 = 0$, Equation (7) holds if, and only if, $P_{k,h}$ is equal to zero or one. By using Equation (4), Equation (5) reduces to the more convenient form:

$$S_{n,m} = -\frac{1}{N}\sum_{i,j} n_{ij} \ln n_{ij} + \ln N. \qquad (8)$$

Now we can define the ideal homogeneous distribution as follows: "A distribution of N points is an ideal homogeneous distribution if for any covering lattice of $n \times m$ cells, each cell (i,j) contains the same number of points", i.e.,

$$n_{ij} = const \qquad i,j(i = 1...n, j = 1...m). \qquad (9)$$

The entropy $S^{(o)}_{n,m}$ for an ideal homogeneous distribution of N points is readily calculated since one has

$$n_{ij} = \frac{N}{n \times m} = const. \qquad (10)$$

Thus, one derives from equation (8) and (10):

$$S^{(o)}_{n,m} = \ln(n \times m). \qquad (11)$$

In the space (n,m,S), Equation (11) defines an entropy surface which is depicted in Figure 2. For an ideal homogeneous distribution, the entropy surface is perfectly smooth. This is no longer the case when deviations occur from the ideal distribution. This can readily be seen in Figure 3 which depicts the entropy surface for the random distribution of 200 points of Figure 1. In Figure 4, the same 200 points have been located in the upper right hand corner of the rectangular grid. Figure 5 shows the roughness of the corresponding entropy surface.
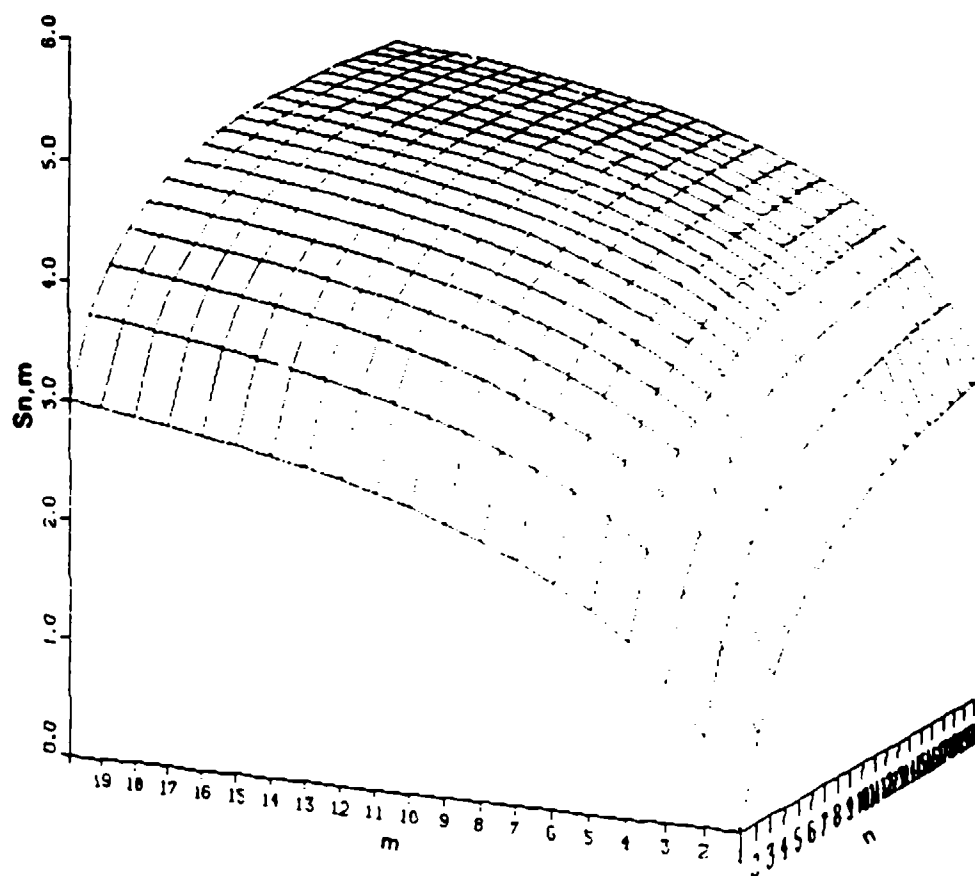
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FIGURE 2. ENTROPY SURFACE FOR AN IDEAL DISTRIBUTION OF 200 POINTS
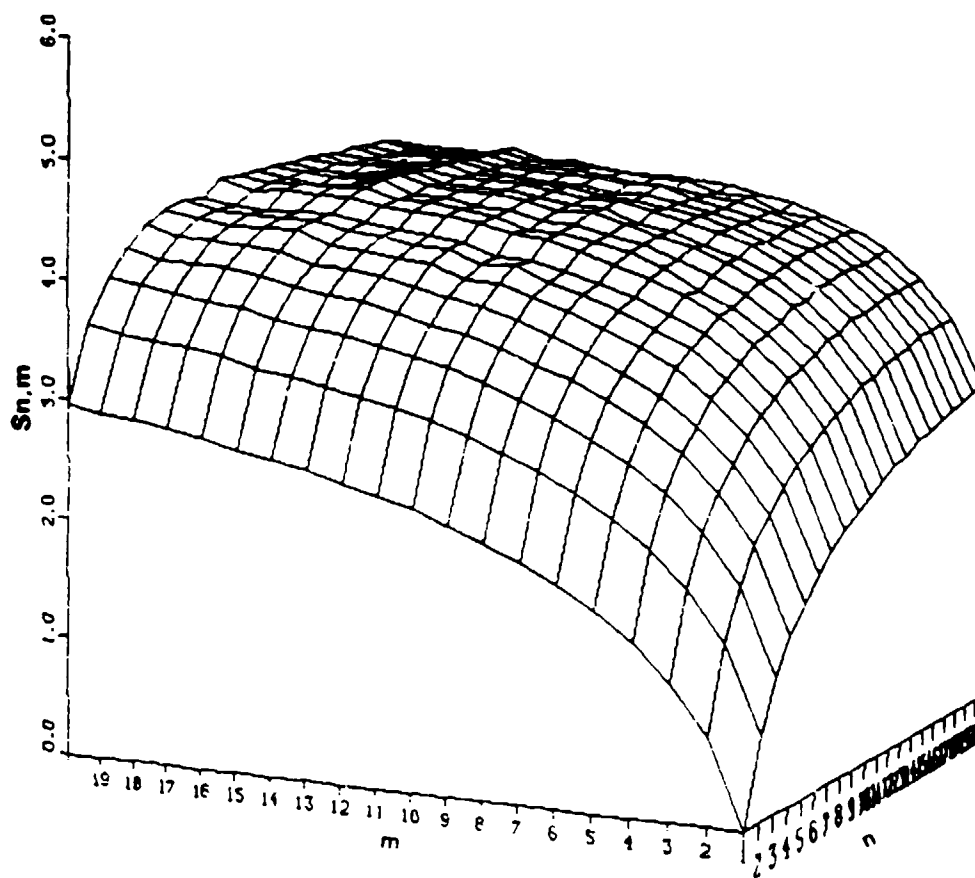
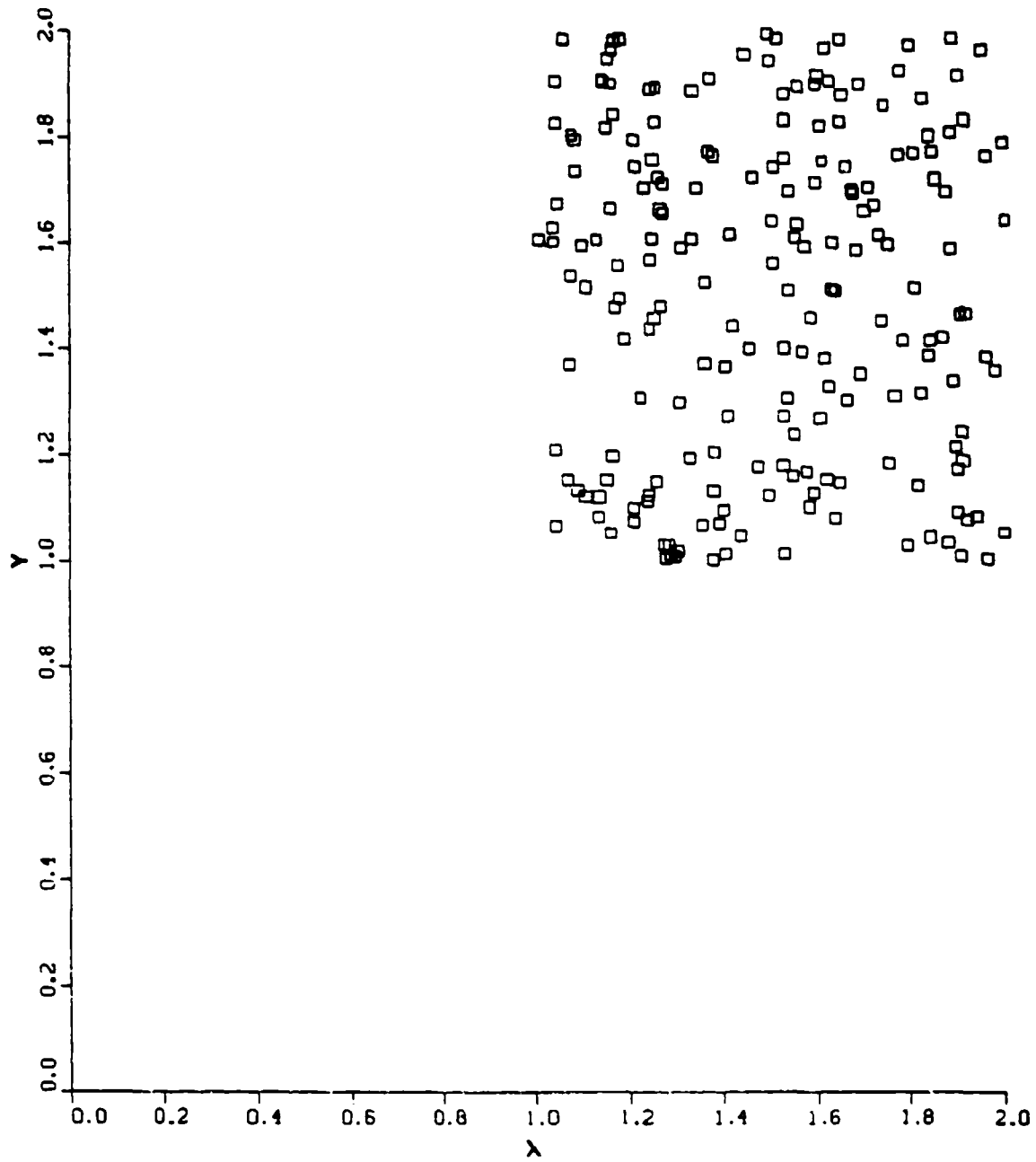FIGURE 3. ENTROPY SURFACE FOR A RANDOM DISTRIBUTION OF 200 POINTS

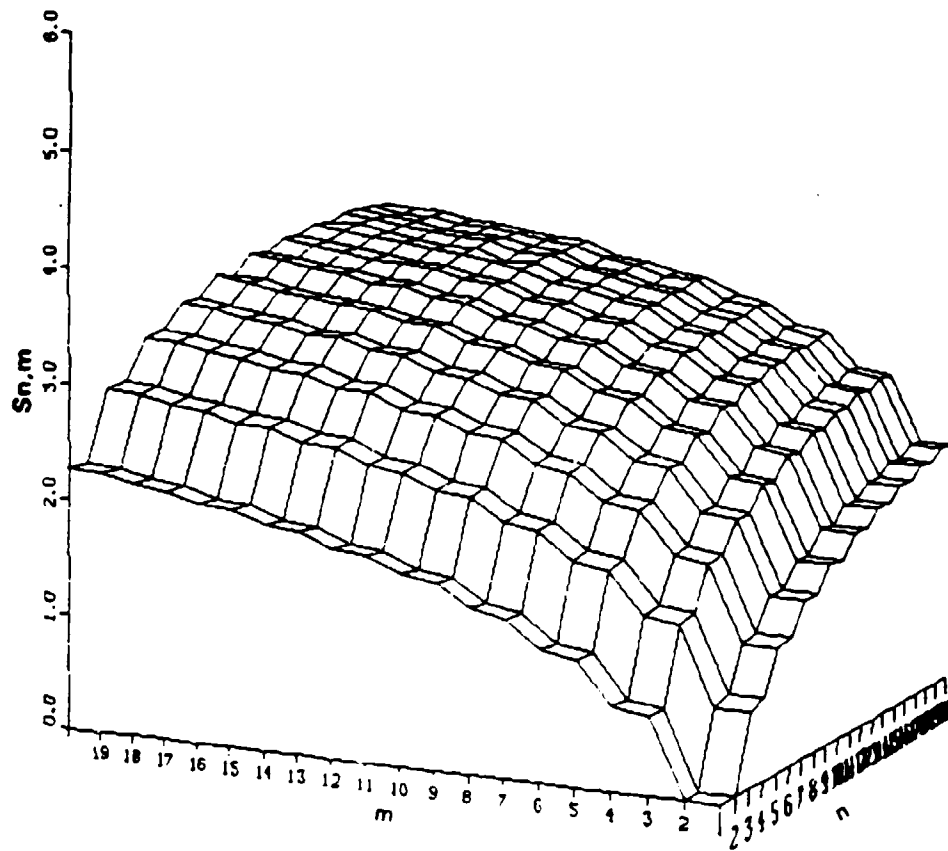FIGURE 4. EXTREME INHOMOGENEOUS DISTRIBUTION OF 200 POINTS

FIGURE 5. ENTROPY SURFACE FOR AN EXTREME INHOMOGENEOUS
DISTRIBUTION OF 200 POINTS

We have shown for an ideal homogeneous distribution that the entropy $S^{(0)}_{n,m}$ does not depend on the number of points but only on the number of cells. It must be pointed out, however, that this is not generally true. By increasing the number of cells, a point is reached at which each cell contains only one point, i.e., the number of cells is equal to the number of points:

$$n \times m - N \qquad (12)$$

When this stage has been reached, any further increase in n and/or m has only the effect of introducing empty cells. Since the partial entropy of an empty cell is zero, the maximum entropy is reached when condition (12) is satisfied. Correspondingly, if $n_{i,j} - 1$ the first term of the right side of Equation (8) goes to zero and the maximum entropy is given by:

$$S^{(0)}_{n,m/max} - \ln N \qquad (13)$$

This is true for any point distribution. In fact, it is always possible to find a lattice fine enough so that each cell contains at most, one point. In other words, for any distribution of points, the entropy reaches a maximum given by:

$$S_{n,m/max} - \ln N. \qquad (14)$$

It must be mentioned that $S_{n,m/max}$ requires a larger number of cells than $S^{(0)}_{n,m/max}$. Since the square is the most symmetrical parallelogram, we select square samples and square-cell lattices, i.e., $n - m$. The lattice

which satisfies condition (12) and the relation:

$$n - m - \lceil \sqrt{N} \rceil \qquad (15)$$

will be called "comparison lattice", whereby $\lceil \sqrt{N} \rceil$ defines the smallest integer $\geq \sqrt{N}$.

ΔS A MEASURE OF INHOMOGENEITY

As a measure of inhomogeneity, we define:

$$\Delta S - S^{(0)}_{n-m/max}(N) \cdot S_{n-m}(N), \qquad (16)$$

whereby $S(N)$ is calculated for the comparison lattice in accordance with Equations (12) and (15).

By combining Equations (8) and (13) one derives:

$$\Delta S - \frac{1}{N} \sum_{i,j-1}^{n} n_{ij} \ln n_{ij} \qquad (17)$$

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ΔS represents a measure of inhomogeneity based upon the comparison lattice which is determined by the total number of particles of a component present in the rectangular sample layer. As discussed above, the entropy S is a function of N, the total number of particles in a sample layer and the number of cells. In Figures 6 and 7, the entropy curves $S^{(o)}{}_{n=m}$ and $S_{n=m}$ are plotted as a function of the number of cells for $N = 200$. The corresponding comparison lattice is calculated from relation (15):

$$n = \lceil \sqrt{200} \rceil = 15$$

Figures 6 and 7 show that the entropy, $S^{(o)}$, for the ideal homogeneous distribution reaches a plateau at values $n \geq 15$. The entropy curves S of Figures 6 and 7 represent the intersections of the entropy surfaces shown in Figures 3 and 5 for square cells, i.e., $n = m$. The corresponding point distributions are displayed in Figures 1 and 4. The ΔS functions reach a maximum in the vicinity of $n = 15$.

ENTROPY OF A SAMPLE BATCH

The previous analysis permits a quantitative evaluation of the degree of deviation of an actual particle distribution from an ideal distribution. The calculation of ΔS requires that the number of particles and reference points be equal. If, however, several samples are taken from a given batch, the values of ΔS, calculated in accordance with Equation (17), provide inaccurate information. This is because each sample contains, in general, a different number of particles and the ΔS's would be inconsistent. Since it is desirable that the number of particles in the test samples and in the reference distribution are equal, a method for calculating ΔS for a sample batch was developed which can be used to adjust the number of particles of the sample batch to the number of particles of the reference distribution.

Assume K^2 samples are taken at random from a given batch; whereby K is a positive integer defined by a given test plan. The samples are squares having sides of a specified length. The K^2 samples are randomized and arranged in a $K \times K$ matrix. Then, the matrix is analyzed with the same algorithm developed in the previous section, i.e., ΔS will be calculated for the $K \times K$ matrix in accordance with Equation (17). A typical calculation is presented in Figures 8 through 12.

CONCLUSION

This paper describes an algorithm, based on the concept of entropy, for the evaluation of homogeneity of a dispersion of particles in a solid mixture. The algorithm is ideally suited for modern computer assisted image analysis systems in quality control of solid mixtures. The application of this algorithm for quality control analysis based on specific selection criteria will be the subject of a forthcoming paper.
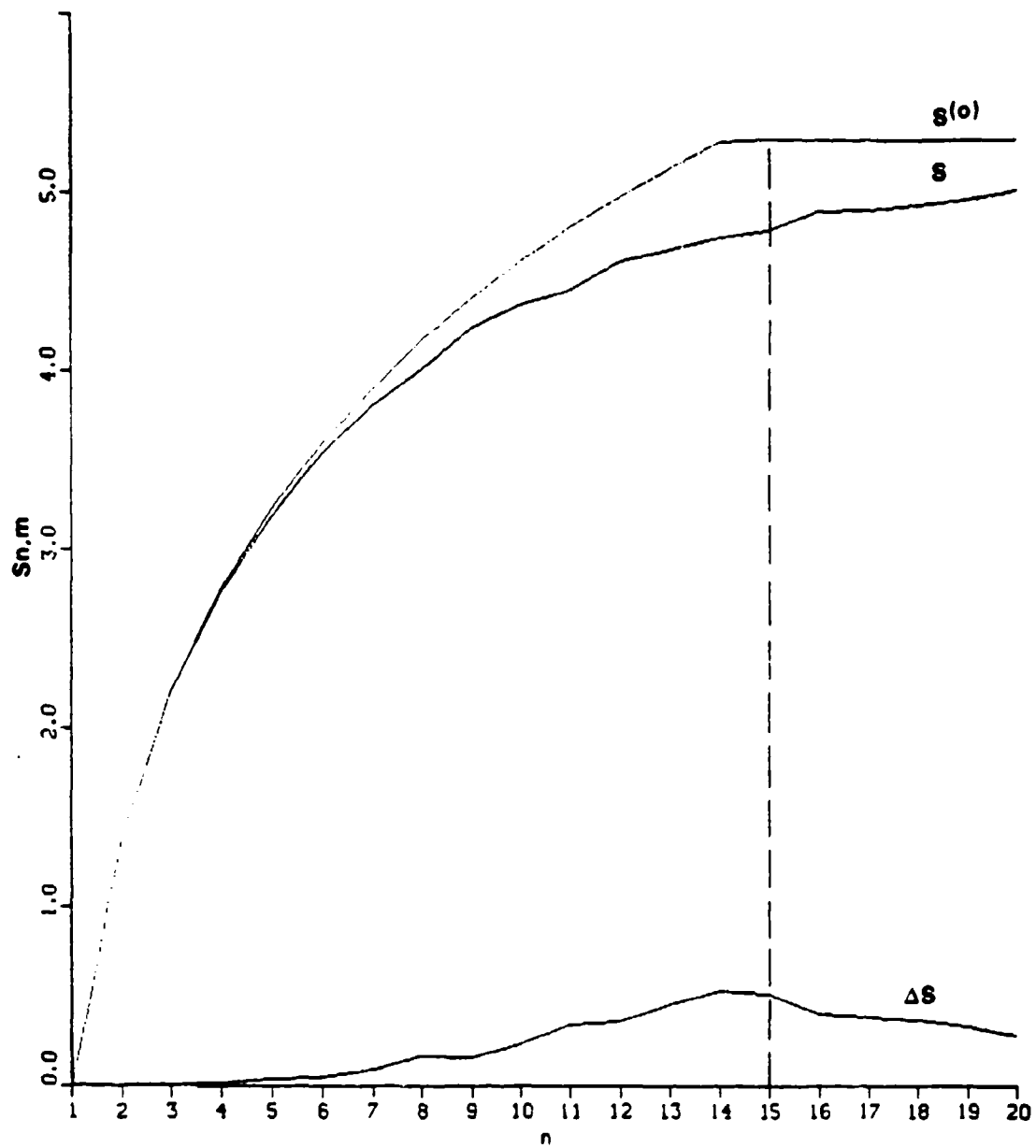
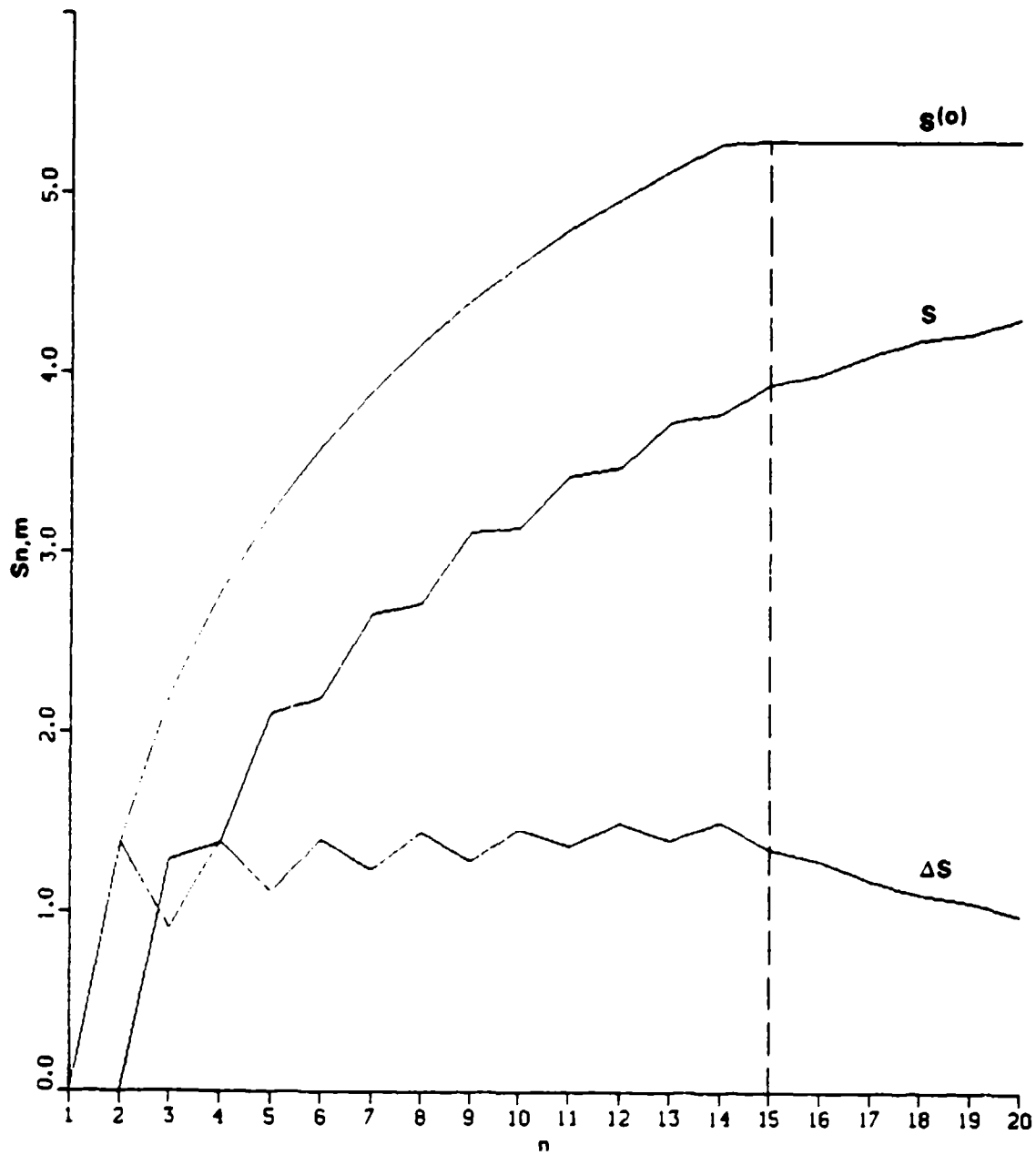FIGURE 6. ENTROPY VERSUS n WHEN n = m FOR A RANDOM DISTRIBUTION OF
200 POINTS

FIGURE 7. ENTROPY VERSUS n WHEN n = m FOR AN EXTREME INHOMOGENEOUS
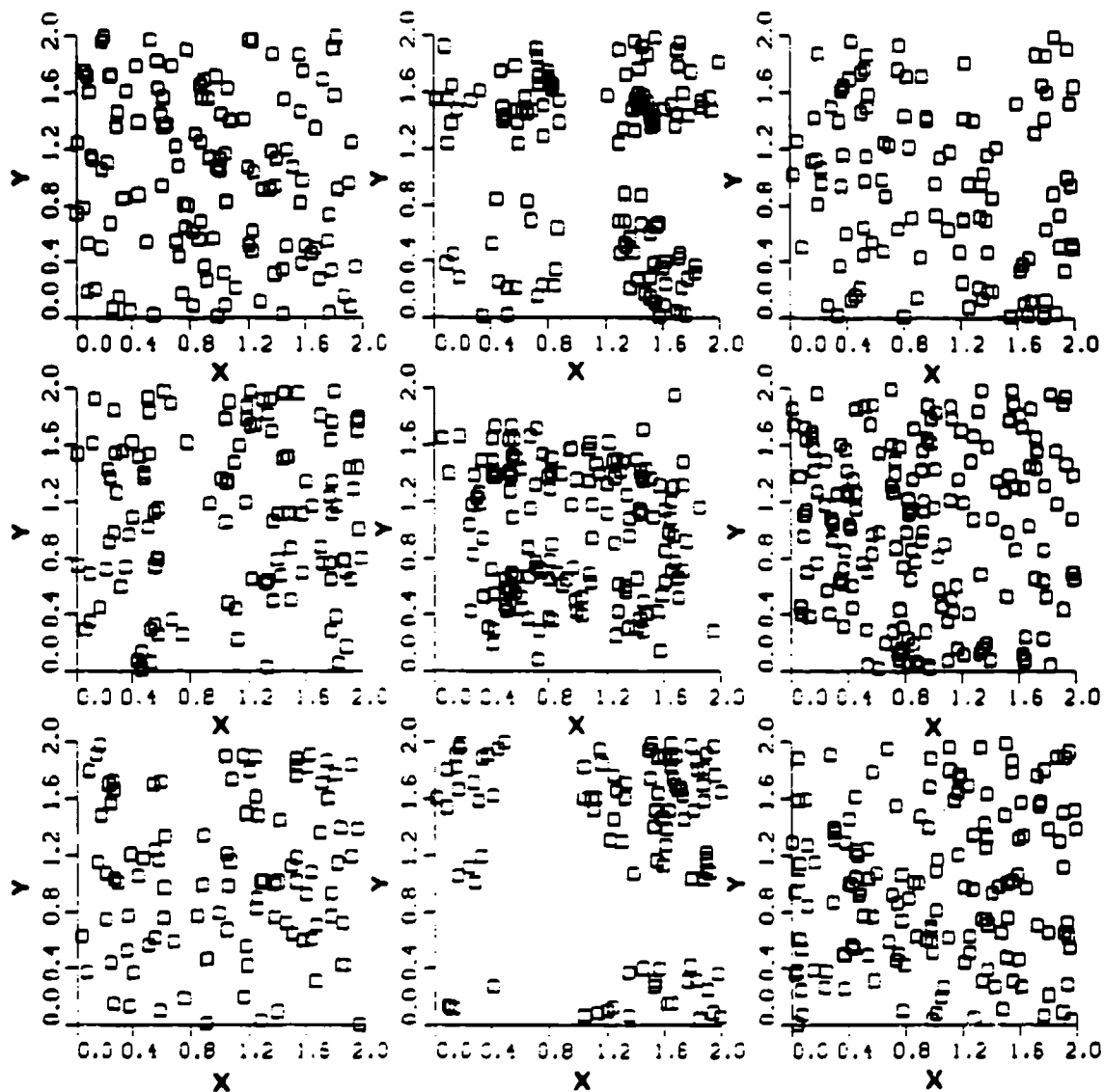DISTRIBUTION OF 200 POINTS
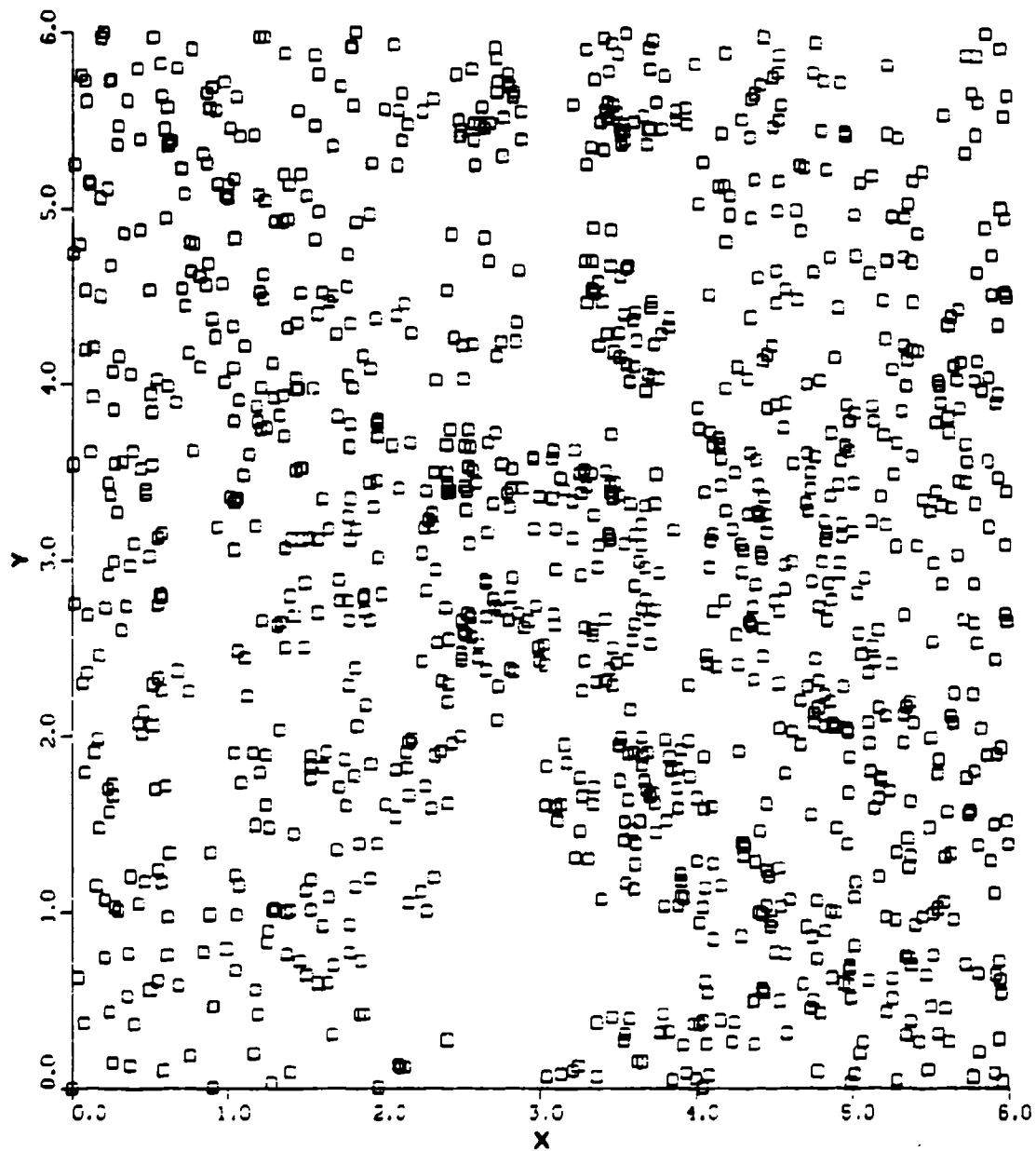
FIGURE 8. POINT DISTRIBUTION IN NINE 2x2 SAMPLES

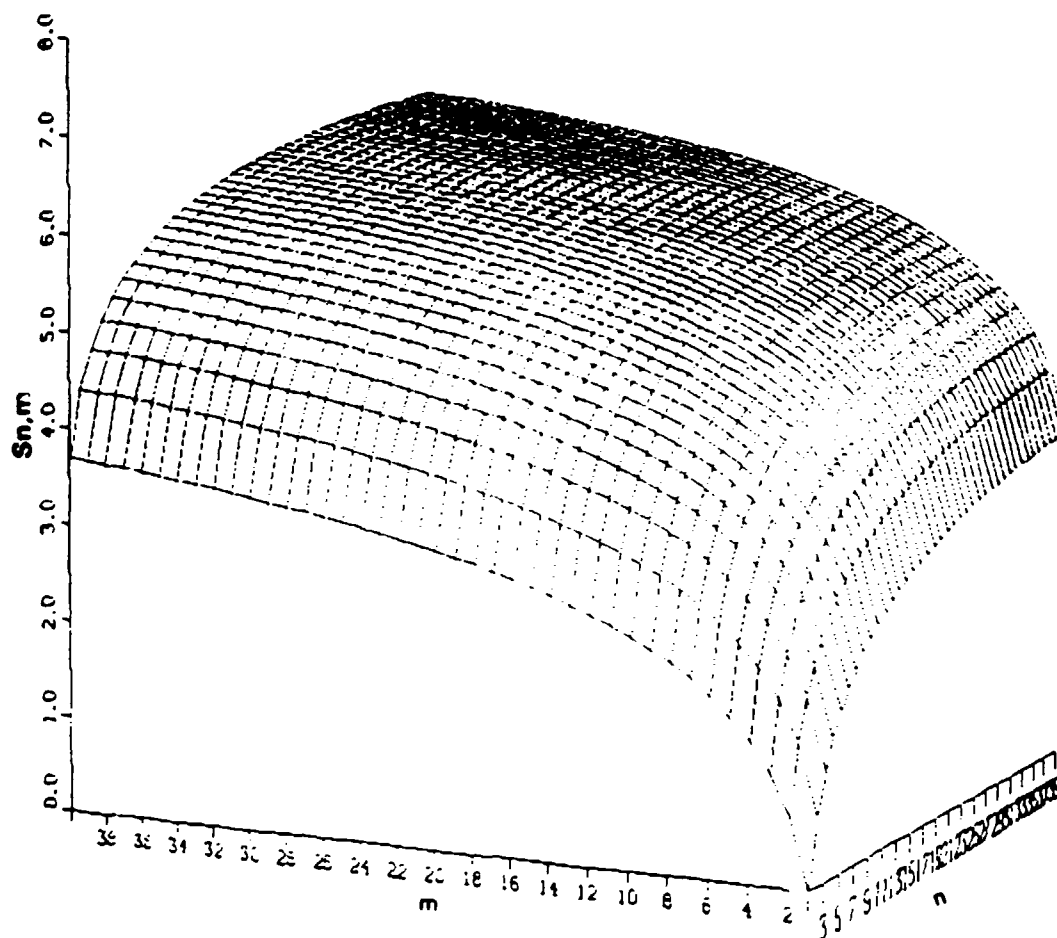FIGURE 9. TRANSLOCATION OF POINTS FROM THE NINE SAMPLES INTO A 6x6 MATRIX

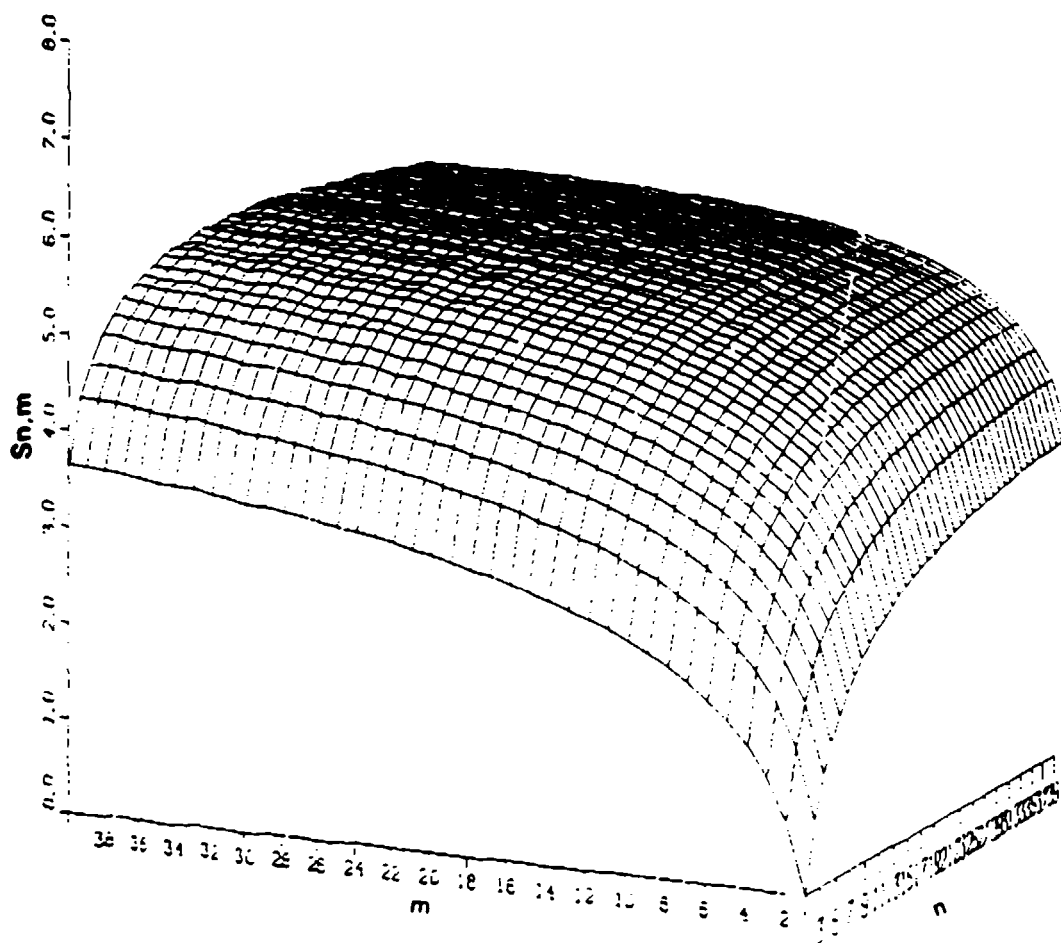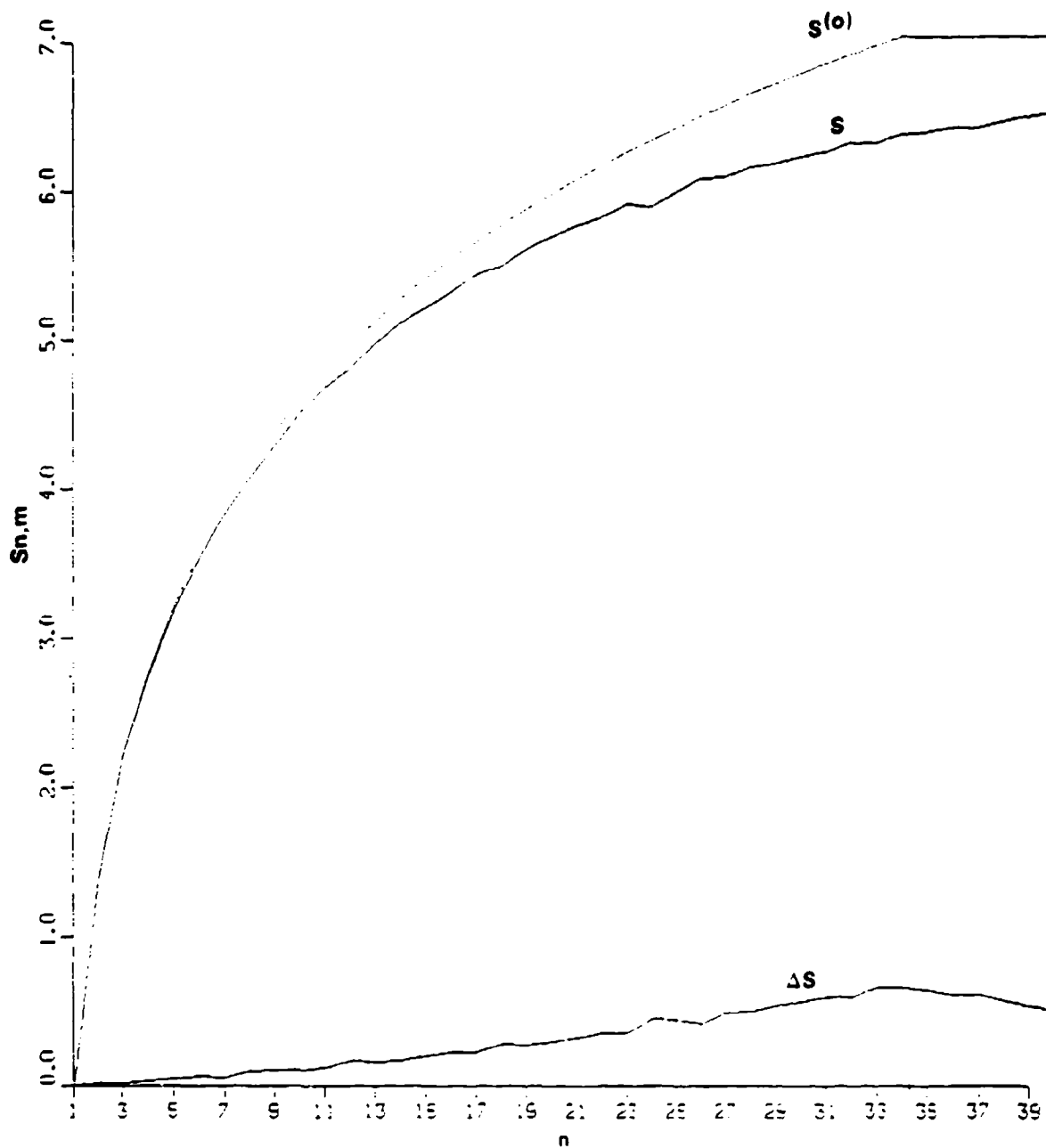FIGURE 10. ENTROPY SURFACE FOR AN IDEAL DISTRIBUTION IN THE NINE SAMPLES

FIGURE 11. ENTROPY SURFACE FOR THE ANALYSIS OF NINE SAMPLES

FIGURE 12. ENTROPY VERSUS n WHEN n = m FOR THE ANALYSIS OF NINE SAMPLES

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